Three-dimensional phase-field simulations of directional solidification

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The phase-field method has become in recent years the method of choice for simulating microstructural pattern formation during solidification. One of its main advantages is that time-dependent three-dimensional simulations become feasible. This makes it possible to address long-standing questions of pattern stability. Here, we investigate the stability of hexagonal cells and eutectic lamellae. For cells, it is shown that the geometry of the relevant instability modes is determined by the symmetry of the steady-state pattern, and that the stability limits strongly depend on the strength of the crystalline anisotropy, as was previously found in two dimensions. For eutectics, preliminary investigations of lamella breakup instabilities are presented. The latter are carried out with a newly developed phase-field model of two-phase solidification which offers superior convergence properties.

I. INTRODUCTION

Microstructural pattern formation during solidification has been intensely investigated for decades [1], and yet progress remains to be made on many fascinating questions. A central one is pattern selection: For a given material under given processing conditions, what is the microstructural pattern that will emerge, and how is it selected out of all other possible structures? This is an important issue both from an applied and a fundamental viewpoint. For the physicist, solidification microstructures are a classic example of pattern formation out of equilibrium, and one would like to have a fundamental principle that is capable to predict the final patterns, as the minimization of an appropriate thermodynamic potential does for equilibrium situations. However, as of yet, no such general selection principle is known for out of equilibrium situations. While empirical rules for solidification microstructures, such as the maximum growth speed or the minimum undercooling criteria, are often very useful in practice, it is often unclear by what detailed mechanisms the final pattern is attained *dynamically*. Therefore, it is important to understand pattern selection more thoroughly starting from the equations for the *evolution* of the solidification front.

Recently, modern methods of statistical physics have allowed theorists to make considerable progress along this line. In particular, the phase-field method has emerged as a powerful tool to simulate microstructure evolution. Its main advantage over more traditional methods for front motion is that it avoids an explicit tracking of the interfaces with the help of one or several *phase fields*, continuous auxiliary fields which distinguish between thermodynamic phases. This makes it possible to simulate the full dynamics of complex morphologies both in two and three dimensions. While up to the mid-1990s, this method had remained a qualitative tool, recent progress has made it possible to treat *quantitatively* some occurrences of the classic free-boundary problem of solidification [2, 3, 4] and to match the results with theory and experiments [5].

Here, we present three-dimensional phase-field simulations of the directional solidification of dilute and eutectic binary alloys. Both cases are characterized by the existence of a continuous family of periodic steady-state solutions with different spatial periodicity (spacing): cells for dilute, lamellae or rods for eutectic alloys. It has been demonstrated both in thin-sample experiments [6, 7] and two-dimensional simulations [8, 9, 10] that no sharp pattern selection occurs: the final spacing depends on the growth history, and a range of stable spacings is observed. Outside of this range of spacings, steady-state solutions generally exist, but they are *unstable*, which explains why they cannot be *dynamically* selected. Therefore, the limits of stability determine the observable spacings. The different instabilities and their onsets have been determined numerically for periodic two-dimensional states [8, 9]; we extend this work to three dimensions, where new instabilities can arise.

II. DILUTE BINARY ALLOY

Thin-sample directional solidification offers a unique opportunity to study the influence of crystalline anisotropy on the interface dynamics experimentally. The anisotropy in the sample plane is determined by the orientation of the three-dimensional crystal with respect to that plane. If a crystalline (100) axis is normal to the sample plane, the in-plane anisotropy is unchanged with respect to the three-dimensional situation; in contrast, if the crystal is oriented with a (111) axis normal to that plane, it is close to zero. It was observed that shallow cells are stable only

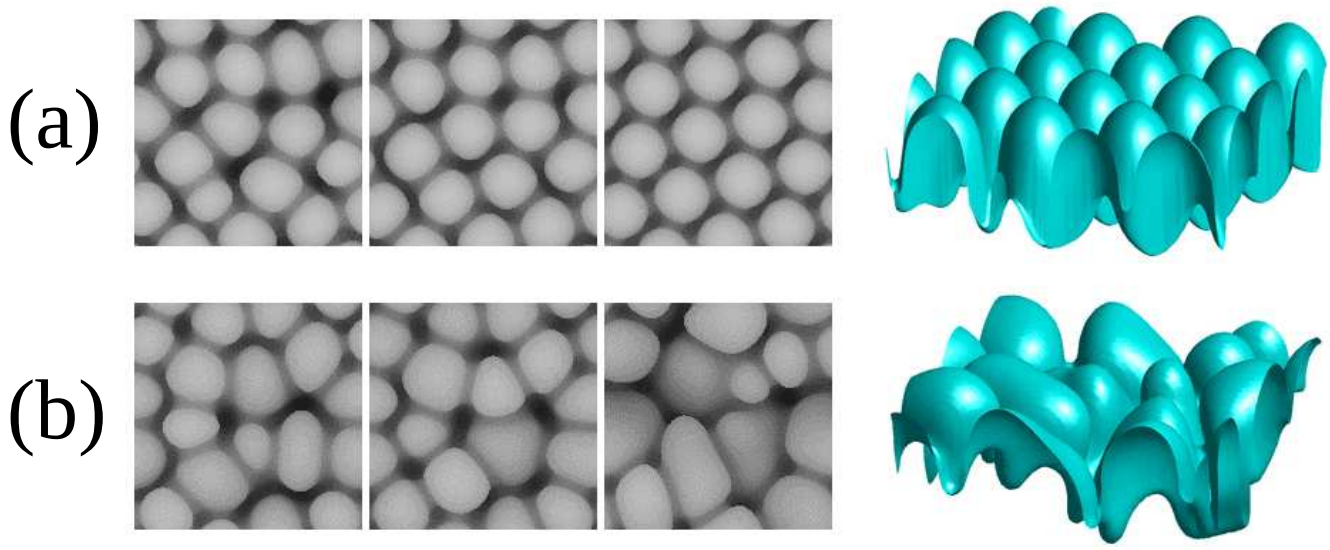

FIG. 1: Evolution of cellular patterns for different anisotropies and otherwise identical parameters: $d_0/l_D = 2.5 \times 10^{-2}$, $\nu = l_T/l_D = 12.5$, where $d_0 = \Gamma/(m\Delta c)$ is the chemical capillary length, $l_D = D/V$ is the diffusion length, and $l_T = m\Delta c/G$ is the thermal length, with $\Gamma, m, \Delta c, D, V, G$ being the Gibbs-Thomson constant, the liquidus slope, the concentration jump (assumed constant), the impurity diffusion coefficient, the growth rate, and the temperature gradient, respectively. (a) $\epsilon_4 = 0.03$ and (b) $\epsilon_4 = 0.02$, where ϵ_4 is the strength of the cubic anisotropy. Left: top views of the growth front (the greyscale is proportional to the surface height); right: 3D view of the interface at the end of the run.

for anisotropic crystals; in the absence of anisotropy, cell splitting and elimination events occur continuously, and the front as a whole never reaches a steady state [6]. This is in agreement with a numerical study in two dimensions by the boundary integral technique [8], which indicated that the range of stable cellular states strongly depends on anisotropy. In addition, this study identified the relevant instability modes that limit the stable range: elimination of every other cell on the short-spacing side, and a period-doubling oscillatory mode on the large-spacing side.

We have extended this investigation to three dimensions using a phase-field model that has equal diffusivities in both phases, and a constant concentration jump (partition coefficient $k = 1$). While this is not very realistic for alloys, the qualitative behavior is similar for a one-sided model [3], and the implementation [11] is particularly simple. Simulations that started from a flat steady-state interface with a small random perturbation are shown in Fig. 1. Clearly, the crystalline anisotropy has a dramatic effect: whereas the interface spontaneously organizes into an array of hexagonal cells for sufficiently strong anisotropy, the evolution remains unsteady up to the end of the run for only slightly lower anisotropy.

To determine the stability boundaries for regular arrays, we generated states with well-defined spacings by placing six cells in simulation boxes of appropriate size and aspect ratio, and changing control parameters until an instability occurred. It turns out that the spatial structure of the relevant instability modes is determined by the symmetry of the underlying steady-state pattern. A hexagonal lattice can be decomposed into three equivalent sublattices of $\sqrt{3}$ times larger spacing. Instabilities occur when the symmetry between the three sublattices is broken. For small spacings, one out of the three sublattices is eliminated. For large spacings, the sublattices start to oscillate, with a phase difference of about $2\pi/3$ between each other. In both cases, once the first cell eliminations or splittings occur, the system follows an unsteady evolution as in Fig. 1b. The stability boundaries are plotted in Fig. 2. The stability limit of the oscillatory mode shifts to larger spacings with increasing anisotropy. This explains the results of the larger simulations in Fig. 1: Only when a sufficiently large range of stable spacings is available, the system is able to "find" a steady state within a reasonable simulation time.

III. EUTECTIC ALLOY

Coupled eutectic growth leads to lamellar or fibrous microstructures. Detailed studies for lamellae in two dimensions by thin-sample experiments [7] and by dynamic boundary-integral simulations [9] have shown that the

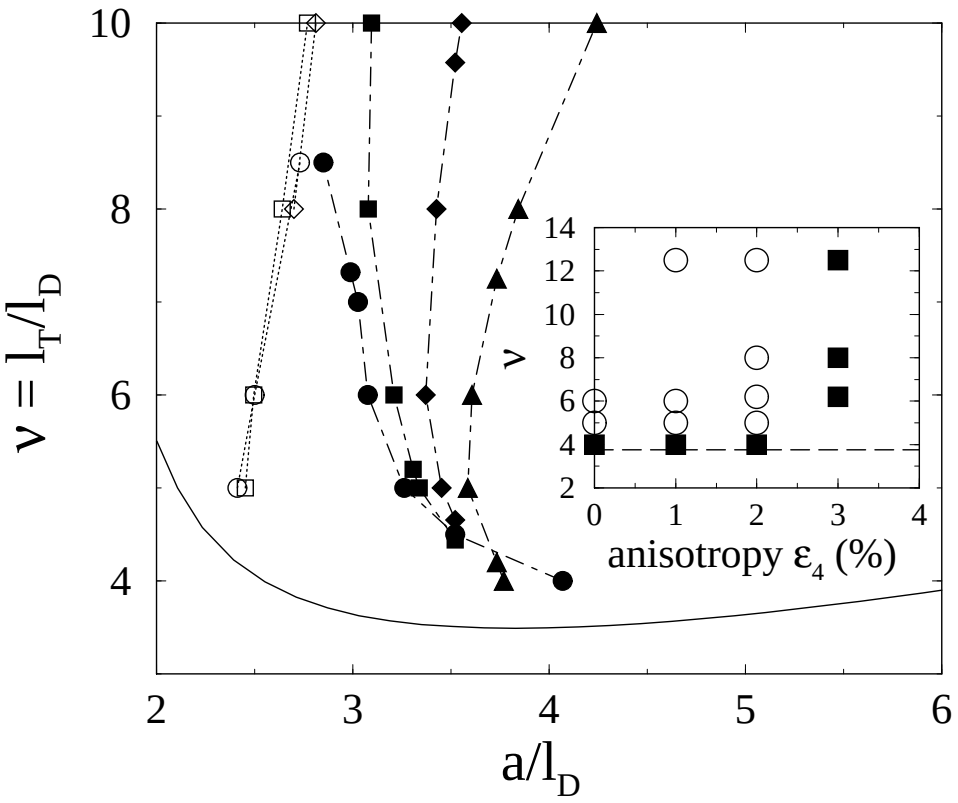

FIG. 2: Stability boundaries of hexagonal arrays of spacing a for different anisotropies: 0% (circles), 1% (squares) 2% (diamonds), and 3% (triangles). Arrays are stable between the limits of cell elimination (dotted lines and open symbols) and oscillations (dash-dotted lines and full symbols). The full line is the neutral stability line of the Mullins-Sekerka instability. Inset: Behavior for runs as those in Fig. 1. Filled squares: stable hexagons; open circles: unsteady evolution; dashed line: Mullins-Sekerka instability threshold.

range of stable spacings is limited on the short-spacing side by a long-wavelength lamella elimination instability, and on the large-spacing side by oscillatory instabilities of once or twice the initial spacing. In contrast, very little is known about pattern stability in three dimensions, since no reliable numerical method was available that could handle complex composite microstructures.

The phase-field method would seem ideally suited to overcome this difficulty. However, its use has been hampered until recently by the wide range of length scales involved, from an interface which is diffuse on a nanometric scale up to a diffusion layer of several millimeters for slow solidification. The problem is that the variation of the phase fields through the diffuse interfaces has to be resolved by the discretization, which would result in huge simulation boxes. Fortunately, *quantitative* phase-field models offer the possibility to use an interface thickness W much larger than that of real solidification fronts, by ensuring that results are independent of W. The only limitation is then that W has to remain about an order of magnitude smaller than the radius of curvature of the interface [2, 3].

We have recently developed a quantitative model for two-phase solidification with vanishing solute diffusion in the two solid phases [4]. As a test, we have performed simulations in two dimensions at the eutectic composition of a model alloy that has a completely symmetric phase diagram (generalization to other compositions and arbitrary phase diagrams is straightforward [4]), and compared the results to a boundary integral calculation with the code of Ref. [9]. As can be seen in Fig. 3, our model produces precise results that are independent of the interface thickness for $\lambda/W \geq 64$, where λ is the lamellar spacing, whereas those of a generic model make appreciable errors and depend on W.

In Fig. 4, we show a three-dimensional simulation of an unstable lamella with reflecting boundary conditions on all sides. The lateral dimensions are 56×168 grid points, and the diffusion field along the growth direction is calculated using a multi–grid method with coarser and coarser grids away from the interface. Undulations develop with time, and finally the lamella breaks up, leaving behind a maze of the two phases (not shown). This evolution can be further

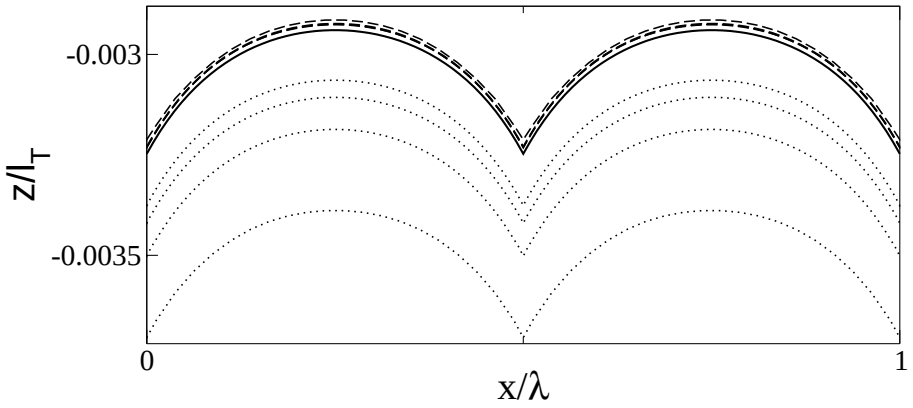

FIG. 3: Benchmark simulations of a two-dimensional lamella pair of spacing λ for $d_0/l_D = 1.95 \times 10^{-5}$, $l_T/l_D = 4$, $\lambda/\lambda_m = 1$, where λ_m is the minimum-undercooling spacing. Solid line: boundary integral. Dotted lines: phase-field model of Ref. [10]. Dashed lines: present phase-field model. Four curves at $\lambda/W = 32$, 64, 96 and 128 shown per model; the results depend on W for the generic model, but superimpose for $\lambda/W \geq 64$ for our new model.

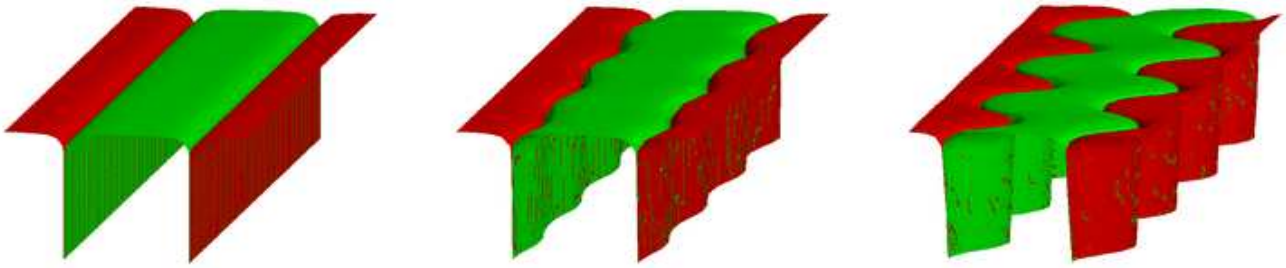

FIG. 4: Snapshot pictures of a three-dimensional lamella that undergoes breakup instabilities. Parameters: $d_0/l_D = 10^{-3}$, $l_T/l_D = 4$, $\lambda/\lambda_m = 1.6$.

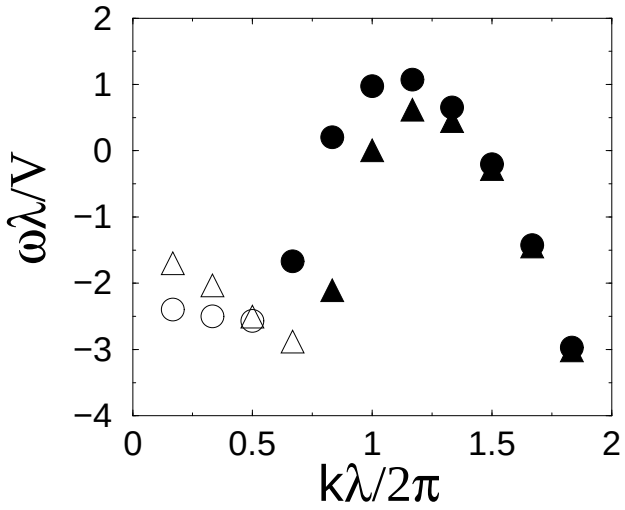

FIG. 5: Stability spectrum (growth rate ω versus wave number k) of the run shown above. Circles: modulations of the mid-line position; triangles: modulations of the lamella thickness. Open symbols: oscillatory modes; filled symbols: exponential modes.

analyzed to characterize the instability. First, the positions of the triple lines are extracted. Then, we calculate the position of the mid-line and the local width of the lamella. These two variables correspond to two different instability modes: the former to an undulation of the whole lamella, the latter to a pinching mechanism. After a Fourier transform, it can be checked that modes of different wave number k along the lamella grow exponentially with a k-dependent growth rate $\omega(k)$. The extracted stability spectrum is plotted in Fig. 5. The "undulating" instability grows faster, and indeed it is this mode that shows up in Fig. 4. It should be noted that this three-dimensional instability occurs for parameters where the two-dimensional system is *stable* both in simulations and experiments. Therefore, it can already be concluded from this preliminary simulation that the range of stable spacings is considerably smaller in three dimensions than in two.

IV. CONCLUSION

Phase-field modeling offers a unique opportunity to investigate pattern stability and pattern selection in three dimensions. Here, we have focused on results for generic phase diagrams, but the method can be carried over to the study of particular materials; simulations for the alloy of Refs. [6, 7] are currently in progress. This will enable us to carry out critical comparisons of simulations, experiments, and theory, which constitutes a particularly promising way to elucidate many long-standing questions concerning microstructural pattern formation during solidification.

V. ACKNOWLEDGMENTS

We thank Alain Karma for many useful discussions, and for the boundary integral code. This work was supported by the Centre National d'Etudes Spatiales, France.

[1] W. Kurz and D. J. Fisher, *Fundamentals of Solidification* (Aedermannsdorf, Switzerland: Trans Tech, 1992); W. J. Boettinger, S. R. Coriell, A. L. Greer, A. Karma, W. Kurz, M. Rappaz, and R. Trivedi, "Solidification microstructures: recent developments, future directions", *Acta Materialia*, 48 (2000), 43-70.

[2] A. Karma and W.-J. Rappel, "Quantitative phase-field modeling of dendritic growth in two and three dimensions", *Phys. Rev. E*, 57 (1998), 4323-4349.

[3] A. Karma, "Phase-field formulation for quantitative modeling of alloy solidification", *Phys. Rev. Lett.*, 87 (2001), 115701.

[4] R. Folch and M. Plapp, "Towards a quantitative phase-field modeling of two-phase solidification", *Phys. Rev. E*, 68 (2003), 010602R.

[5] A. Karma, Y. H. Lee, and M. Plapp, "Three-dimensional dendrite tip morphology at low undercooling", *Phys. Rev. E*, 61 (2000), 3996-4006.

[6] S. Akamatsu and G. Faivre, "Anisotropy-driven dynamics of cellular fronts in directional solidification in thin samples", *Phys. Rev. E*, 58 (1998), 3302-3315.

[7] M. Ginibre, S. Akamatsu, and G. Faivre, "Experimental determination of the stability diagram of a lamellar eutectic growth front", *Phys. Rev. E*, 56 (1997), 780-796.

[8] P. Kopczynski, W.-J. Rappel, and A. Karma, "Critical role of crystalline anisotropy in the stability of cellular array structures", *Phys. Rev. Lett.*, 77 (1996), 3387-3390.

[9] A. Karma and A. Sarkissian, "Morphological instabilities of lamellar eutectics", *Met. Mat. Trans. A*, 27 (1996), 635-656.

[10] R. Folch and M. Plapp, "Phase-field modeling of eutectic solidification: From oscillations to invasion", *Interface and Transport Dynamics*, ed. H. Emmerich, B. Nestler, and M. Schreckenberg (Springer, Berlin, 2003), 182-189; preprint cond-mat/0206237.

[11] M. Plapp and M. Dejmek, "Stability of hexagonal solidification patterns", Europhys. Lett. 65 (2004), 276-282; preprint cond-mat/0207314.